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ULTRASOUND AS AN EMERGING TECHNOLOGY FOR THE ELIMINATION OF CHEMICAL CONTAMINANTS IN FOOD: A REVIEW

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Abstract: Background: The existence of chemical contaminants in food brings a serious threat to human health. Researchers have been making persistent efforts to eliminate contaminants from food to make it safe for human consumption. Traditional methods, such as washing with various agents, peeling, cooking and chemical oxidants, cannot achieve the desired results. In recent years, non-thermal technology, such as ultrasound, has attracted extensive attention as an emerging technology for removing food contaminants. Scope and approach: This review analyzed the research status of ultrasound in the elimination of food contaminants, studied the transformation products and their potential toxicity during ultrasound treatment, discussed the effect of ultrasound on food quality, and summarized the advantages and disadvantages of ultrasound treatment. At the same time, the challenges of this technology, the problems to be studied, and future development prospects were discussed

Key findings and conclusions: Ultrasound is a green processing technology, which would not impart secondary pollution. It also has advantages of high efficiency and low energy consumption, compared with traditional decontamination technologies. The combination of ultrasound with other techniques such as ultraviolet, ozone and pulsed electric field shows better decontamination effect. However, ultrasound treatment may cause degradation of some phenolic compounds and vitamins, changes in color, loss of anthocyanin and other adverse effects on food characteristics. In

addition, the appropriate ultrasound processing system in terms of probe design, geometry, and operating conditions, needs to be specially designed for different food materials. Thus, some challenges need to be addressed to improve its application.

1. Introduction

Food safety is a major public health issue related to people's livelihood, economic development and political stability. Chemical contaminants such as pesticides, heavy metals, mycotoxins, and allergens have seriously affected the safety of food consumed according to published reports (Afonne & Ifediba, 2020; Narenderan, Meyyanathan, & Babu, 2020). For example, pesticides are used to protect crops from pests, which can lead to residues on harvested crops and to health concerns among consumers (An et al., 2018). Additionally, environmental issues related to the use of pesticides have been identified as public concerns(Villaverde et al., 2018). Statistical reports revealed that about 3.5 million tons of pesticides are used in global agricultural production every year, of which 70% were in China, the United States and Argentina (Hvezdova et al., 2018). Pesticide residues in the environment may accumulate in human tissues through the food chain, which may harm human health, cause myasthenia, endocrine disorders, respiratory disorders, paralysis, and cancer, among other disorders (Nougadere et al., 2020). With the continuous development of modern industry, heavy metal pollution is becoming more and more serious, causing many public problems (Rahman & Singh, 2019). Heavy metals are proven to be chemically stable and, as a result, human beings are at high risk of an inevitable exposure to these life-threatening heavy metals through food chains. Moreover, the harm of heavy metals poisoning to the human body is proved to be multi-organ, multisystem, multi-indication and irreversible (Jamshidifard et al., 2019). Food allergy refers to an abnormal or exaggerated immune response triggered by eating specific foods or food additives. It is one of the most common disorders and has tended to increase in prevalence in the past decades (Gupta et al., 2019). At present, there are more than 170 kinds of foods or food components that can cause allergic reactions in sensitive people (Khan et al., 2019). These foods can be divided into the following eight categories: milk, eggs, soybeans, wheat, fish, shellfish, tree nuts and peanuts. Food allergy can cause nausea, vomiting, pruritus, shock and death (Van Vlierberghe et al., 2020). However, there is no effective treatment for food allergy, which can only be prevented by avoiding the allergen. Mycotoxins are toxic secondary metabolites produced by some fungi (moulds) (Wielogorska et al., 2019). These low molecular weight compounds (usually less than 1000 Da) are naturally occurring and inevitable. Eating foods or feed contaminated with mycotoxins can cause acute or chronic toxicity in humans and animals (Kebede, Liu, Jin, & Xing, 2020). It causes unpredictable and persistent food safety problems all over the world. Therefore, how to remove the above-mentioned chemical pollutants in food effectively to obtain safe food has become a topic of common concern. Conventional processing technologies, such as washing with various agents, peeling, cooking and chemical oxidants, have been extensively investigated to reduce contamination in foodstuff (Alister et al., 2018), but none of them are completely satisfactory. Such methods cannot effectively remove harmful substances or may cause secondary pollution (Yigit & Velioglu, 2019). Besides, some extreme treatment conditions may affect the sensory quality of food or cause the loss of nutritional components (Yang et al., 2012). Hence, the application of a new technology, such as ultrasound, in the detoxification of food pollution is a current research hotspot. Ultrasound has been widely reported to be able to degrade organic pollutants in the field of environmental science (Debabrata & Sivakumar, 2018; Kida, Ziembowicz, & Koszelnik, 2018).

Applications of ultrasound for food products.

Applications	Conventional methods	Advantages	Products	References
Extraction	Maceration Percolation	Less time Higher yields Lower temperatures	Aromas Polyphenols Minerals	Carrillo-Hormaza, Duque, López-Parra, and Osorio (2020); Umana, Eim, Garau, Rosselló, and Simal (2020); Zamanipoor et al. (2020)
Emulsification	Mechanical treatment	Less time Better stability	Emulsions (ketchup, mayonnaise, ...)	Li, Li, et al. (2020)
Freezing/crystallization	Freezer Freezing by immersion, by contact, ...	Less time Small crystals Improving diffusion Rapid temperature decreasing	Meat Vegetables Fruits Milk products	Tian, Zhang, Zhu, and Sun (2020)
Filtration	Filters (permeable membranes, ...)	Less time Improving filtration	Juices	Aliasghari et al. (2015)
Cooking	Fryer Stove Water bath, ...	Less time Improving heat transfer and organoleptic quality	Meat Vegetables	Miano, Sabadoti, and Augusto (2018)
Drying	Hot gas stream Freezing Microwave	Less time Improving heat transfer and organoleptic quality	Fruits Vegetables	Guo et al. (2020)
Cutting	Knives	Less time Reducing products losses Accurate and repetitive cutting	Fragile products (cake, cheese, ...)	Yildiz, Rababah, and Feng (2016)
Degassing	Mechanical treatment	Less time Improving hygiene	Chocolate Fermented products (Beer, ...)	Chemat, Zill e, and Khan (2011)
Defoaming	Thermal treatment Chemical treatment Electrical treatment Mechanical treatment	Less time Improving hygiene	Carbonated drinks Fermented products (Beer, ...)	Rodríguez et al. (2010)
Demoulding	Greasing moulds Teflon moulds	Less time Reducing products losses	Cooked products (cake, ...)	Chemat et al. (2011)
Fermentation	Microorganisms	Less time Improving rheological properties Improving organoleptic quality	Yoghurt Wine	Dahroud et al. (2016); Zhang, Xu, Chen, Zhao, and Xue (2020)
Oxidation	Contact with air	Less time	Alcohols (wine, whisky, ...)	Del Fresno et al. (2018)
Marinating	Brine	Less time Improving organoleptic quality Product stability	Vegetables Meat Fish Cheese	Shi et al. (2020)
Enzymatic And Microbial Inactivation	Pasteurization Sterilization Sanitizers	Improving organoleptic quality Retaining bioactive substances	Juices Milk Vegetables Fruits	Scudino et al. (2020); Wang, Liu, et al. (2020)

2. Ultrasonic mechanism and system

Ultrasound is defined as a sound wave at a frequency that exceeds the hearing limit of the human ear (20 kHz) (Yu et al., 2020). When acting on a liquid medium, the ultrasound wave generates positive and negative pressure, which produces periodic compression and expansion on the liquid medium molecules. The alternating speed depends on the ultrasonic frequency. In the stage of negative pressure expansion, the ultrasonic wave will produce tiny bubbles or cavities in the liquid medium. These cavities keep absorbing acoustic energy until they reach a critical size and break violently, releasing high energy. This phenomenon is called the cavitation effect, which is favored by the food industries for its excellent cleaning, processing and disinfection performance (Khanal, Anand, Muthukumarappan, & Huegli, 2014). Three types of ultrasound are classified according to the frequency range: power ultrasound (20–100 kHz), high-frequency ultrasound (100 kHz – l MHz) and diagnostic ultrasound (1–10 MHz). For the high- frequency ultrasound and diagnostic ultrasound, they are normally utilized for soft tissue surgery, or diagnostic imaging, increased drug delivery, and simulation of tissue regeneration (Wischhusen & Padilla, 2019). Power ultrasound has been widely studied in the food processing, such as structural modification of different food proteins, efficient extraction of various biological activities, sterilization and freeze-drying (Chen et al., 2020). Stable cavitation is produced in the process of continuous low-intensity ultrasound (<1 W/cm2), the volume of the cavitation bubble gradually expands, but remains smaller than the critical fracture size during the compression cycle. Transient cavitation refers to the rapid expansion of the cavitation bubble in the process of continuous high-intensity ultrasound (>1 W/cm2), which breaks and releases a lot of energy after reaching a critical size (as shown in Fig. 1). Such a unique environment generated by ultrasound can fragment water and other molecules into free radicals. Therefore, power ultrasound may become a potential means of eliminating chemical contaminants in the food industry. An appropriate ultrasonic sensing or processing system is the basis for the successful application of ultrasonic technology in industrial decontamination. Recently, it has been observed that intensive research concerning probe design, geometry, and characteristics (e.g., frequency) as well as operating conditions, are being carried out with the aim to meet the demands of specific applications in different food materials or provide optimum results for each individual application (Clodoveo et al., 2017). As shown in Fig. 2, the ultrasonic probes determine the cavitation phenomenon within the water along with a vibrational energy transferred to the processing food. The heat exchanger can be used to precisely adjust the temperature of the system during the ultrasonic treatment of food. If the food is solid or semi-solid state, it is necessary to select the appropriate ultrasonic medium (such as water). If it is liquid food, it can directly enter the system for ultrasonic treatment.

3. Elimination of food contaminants by ultrasound technology

3.1. Pesticides

Pesticide residues in food, and especially fruits and vegetables, are extremely harmful to human health because most of those items are eaten raw or slightly processed (Bhilwadikar, Pounraj, Manivannan, Rastogi, & Negi, 2019). Therefore, necessary measures are imperative to remove the pesticides from the agricultural produce. In the past decades, several methods such as washing with various agents, peeling, salting, cooking and chemical oxidants were applied to remove pesticides in agriproducts. However, these traditional operations have their limitations. For example, washing in tap water cannot effectively remove pesticides; thermal treatment will lead to the loss of nutrients such as vitamins in fruits and vegetables; the use of a chemical agents is more likely to produce toxic by-products, causing secondary pollution to food (Phan et al., 2018; Yang et al., 2012). As an alternative method, ultrasonic technology has been successfully used to remove pesticide residues in food. Lozowicka, Jankowska, Hrynko, and Kaczynski (2016) reported that ten fungicides and six insecticides in strawberry dissipated significantly after ultrasonic cleaning, with the removal rate ranging from 45.6 to 91.2%. In contrast, the removal rate of fungicides and insecticides after washing with tap water ranged from 19.8% to 68.1%. Similarly, an 84% reduction in the level of fluopyram in apple samples was observed after ultrasonic washing, which was significantly higher than the reduction after washing with tap water (56.2%) and boiling at 100 ◦C (72.1%) (Slowik-Borowiec & Szpyrka, 2020). The principle of ultrasonic cleaning was to destroy the adsorption between pesticides and the surface of fruits and vegetables, thus removing the adhered pesticides from fruits and vegetables. The chemical properties of pesticides can vary greatly, some of which are fat soluble and some water soluble; this must be taken into account when applying a cleaning method. Therefore, the removal process of pesticides varies with pesticides and products.

Table 2
Summary of recent research on pesticides elimination by ultrasound.

Pesticides	Treated sample	Process condition	Maximum degradation rate[a]	Reference
Malathion and chlorpyrifos	Apple juice	pH 3.86, 2–3 mg/L, 25 kHz, 500 W, 120 min	Malathion (42%) and chlorpyrifos (82%)	Zhang et al., 2010
Methamidophos and dichlorvos	Lettuce	Ultrasound/O_3 combination treatment for 60 min (Ultrasound: 25 kHz, 500 W, O_3: flow rate 75 mg/min)	Methamidophos (82%) and dichlorvos (86%)	Fan, Zhang, Xiao, Qiu, and Jiang (2015)
Chlorpyrifos ethyl, τ-fluvalinate, cyprodinil, pyraclostrobin, and malathion	Sour cherry juice	Pulsed electric field (20 kV/cm for 600 μs) + Ultrasound (28 kHz for 30 min)	Ranged from 43 to 93%	Akdemir Evrendilek et al. (2020)
captan, thia-methoxam and metalaxyl	Tomato	Low intensity electrical current (1400 mA) and ultrasound (40 kHz)	Captan (94%), thiamethoxam (70%) and metalaxyl (95%)	Cengiz et al. (2018)
Acephate, malathion, carbaryl, bifenthrin, cypermethrin, permethrin, cyhalothrin, chlorothalonil, and imidacloprid	Tomato	Water washing with ultrasound (400 W)	Acephate (55%), malathion (66%), carbaryl (75%), bifenthrin (68%), cypermethrin (68%), permethrin (60%), cyhalothrin (70%), chlorothalonil (78%) and imidacloprid (72%)	Al-Taher, Chen, Wylie, and Cappozzo (2013)
Ten fungicides and six insecticides	Strawberry	Ultrasonic cleaning (40 kHz, 240 W)	Ranging from 46 to 91%	Lozowicka et al. (2016)
Fluopyram and tebuconazole	Apple	Ultrasonic washing (35 kHz, 240 W)	Fluopyram (84%) and tebuconazole (79%)	Slowik et al. (2020)
Forchlorfenuron	Kiwifruit juice	40 kHz, 600 W, 45 °C for 40 min	72%	Li, Guo, et al. (2020)
Chlorothalonil, pyrazophos, and carbendazim	Pakchoi	28 kHz, 0.45 W/cm^2, 20 °C for 10 min	Chlorothalonil (75%), pyrazophos (46%) and carbendazim (25%)	Zhu et al. (2019)

[a] Some values are approximated.

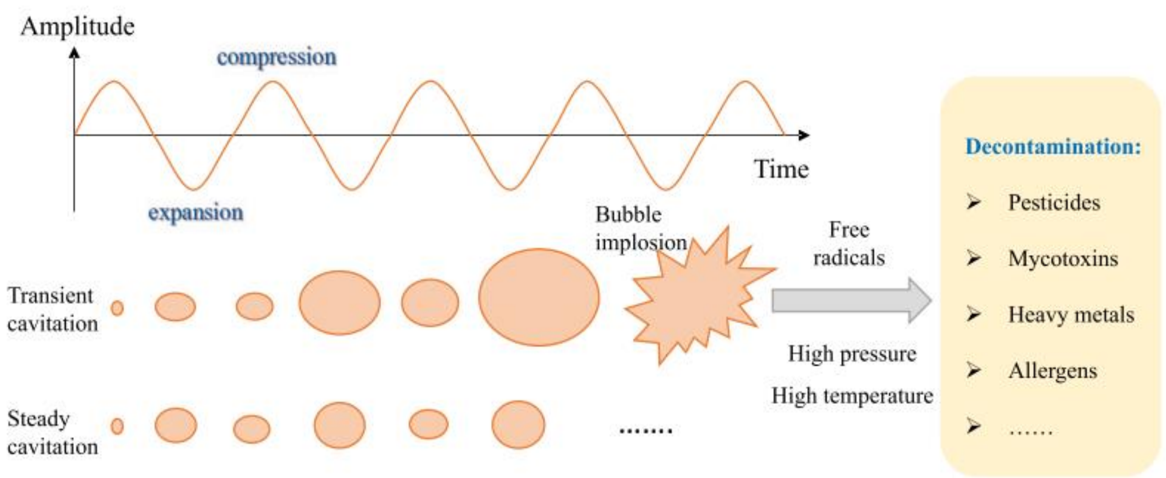

Fig. 1.Cavitation effect and its application in food decontamination.

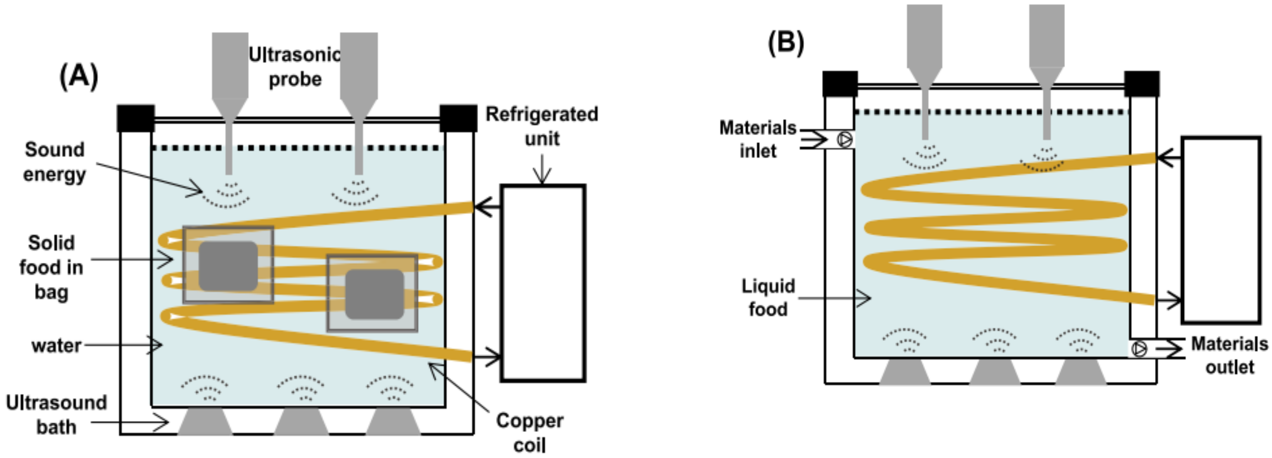

Fig. 2.Schematic diagram of ultrasonic machining device for (A) solid or semi-solid food and (B) liquid food.

3.2. Mycotoxins

Similar to the removal of pesticide residues in food, ultrasound has also been used to reduce mycotoxins in agricultural products. A study showed that thermal ultrasound could reduce 52.07% aflatoxin M1 in milk compared with the control group (Hern´ andez-Falc´ on et al., 2018). The ultrasonic degradation of aflatoxin was pointed out to be related to the opening and hydrolysis of the lactone ring or the rupture of the furan double ring of aflatoxin, which then leads to changes in the molecular structure (Liu, Li, Liu, & Bian, 2019). From a previous review, the aflatoxin reduction by conventional thermal processing showed a rate ranging from 42 to 90%, which seems to be equivalent to the effect of ultrasonic treatment (Kabak, 2009). However, the advantage of ultrasonic treatment is that it does not generate harmful substances, such as acrylamide and advanced glycation end products. For the degradation mechanism of aflatoxin upon ultrasound treatment, Liu et al. (2019) believed that the addition of ·H, ·OH and H2O2 generated by ultrasound led to the hydrolysis and oxidation of aflatoxin molecules. Unlike the direct oxidation of mycotoxins, another study indicated that 75 ◦C thermosonication led to the inactivation of Byssochlamys nivea ascospores in strawberry puree, thus, reducing the possibility of mycotoxin production (Evelyn & Silva, 2015). These studies mentioned above show the potential of ultrasound as an emerging technology to remove mycotoxins from food, ensuring safer foods with high sensory quality. However, there are only a few reports about the effect of ultrasound technology on mycotoxins in the food area; more studies are needed to investigate the mechanisms of ultrasound in the elimination of mycotoxins.

3.3.Allergens

After ultrasonic energy was proved to destroy the tertiary and quaternary structures of casein, and affect the physicochemical and functional properties of whey protein (Villamiel & Jong, 2000), scholars began to study the effect of ultrasound treatment on food allergens (Table 3). In peanut, one study reported that ultrasonic treatments significantly reduced Ara h 1 (99.8%) and Ara h 2 (98.82) levels in peanuts, and resulted in a 50% inhibition on IgE binding (Li, Yu, Ahmedna, & Goktepe, 2013). The reduction in peanut allergens is due to the structural changes of IgE binding epitopes located on the a-helical regions of Ara h 1 and Ara h 2 under the high-intensity ultrasound (HIU) treatment for a long time duration. In shrimp, another study revealed that HIU (100–500 W) degraded the tropomyosin from Exopalaemon modestus to generate protein fragments by HIU-induced free radicals, thus, reducing the allergenicity of tropomyosin. Stronger HIU (600–800 W) could strengthen the allergenicity reduction (Zhang, Zhang, Chen, & Zhou, 2018). However, the effect of ultrasound on allergens in food is not always positive. In milk, it has been reported that ultrasonic treatment of 0–500 W did not reduce the allergenicity of α-casein and whey proteinscan (Tammineedi, Choudhary, Perez-Alvarado, & Watson, 2013), and increased the allergenicity of β-lactoglobulin (Shao, Zhang, Liu, & Tu, 2020). Thus, it can be seen that the effect of ultrasound treatment on the elimination of allergens in milk relies on process parameters such as ultrasonic frequency, intensity, treatment duration and the composition of milk. Allergens are generally sensitive to ultrasound, so ultrasound processing shows a potential application in the reduction of allergens in soy, peanut, shrimp, etc. It is also noticed that ultrasound treatments seem to be more efficient when combined with heat, instead of being used solely. Therefore, ultrasound can be considered as an alternative non-thermal processing in the reduction of food allergens, whereas the optimization of processing conditions is needed (e.g., time duration, frequency, and temperature) (Rahaman, Vasiljevic, & Ramchandran, 2016).

Conclusions

Ultrasound technology is an environmentally friendly technology that is highly efficient and gentle, which not only improves processing efficiency and product quality, but also saves cost. This review discussed the feasibility of eliminating contaminants using ultrasound technology. Despite being in its infancy, the research progress in this subject has already shown that ultrasound processing was effective in removing pesticide residues and allergens in fruits, vegetables and milk. The combination of ultrasound with other techniques such as ultraviolet, ozone, and PEF showed better decontamination effect. In addition, ultrasonic treatment can enhance the color and reduce the loss of volatile components in fruit juice. Besides, it can tenderize meat products, and retain bioactive substances in food. However, the best conditions involving ultrasound frequency, acoustic energy density, time of treatment, and temperature need to be explored to minimize the potential adverse effects on food quality and safety. The appropriate ultrasound processing system in terms of probe design and geometry, needs to be specially designed for different food materials. It can be predicted that after further investigations, ultrasound technology will play a significant role in the approachable techniques of food decontamination.

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